

PATENT ABSTRACTS OF JAPAN

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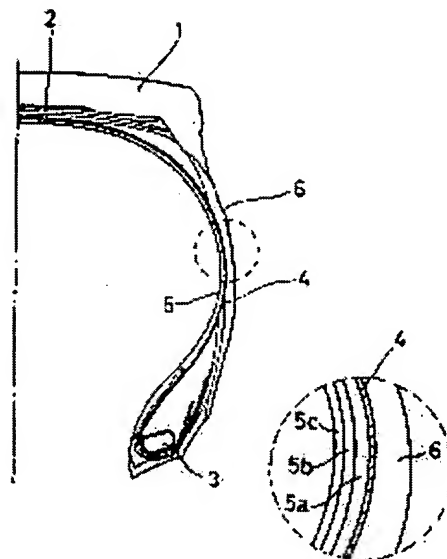
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(54) PNEUMATIC TIRE

(57)Abstract:

PURPOSE: To improve the durability of a tire in an extensive application area ranging from a normal area to a high temperature and humid area by forming an inner liner layer to be set up at the inside of a carcass layer into a three- layer structure, and specifying a rubber composition forming each layer.

CONSTITUTION: A carcass layer 4 is set up between a symmetrical pair of bead wires 3, and in the inner part of this carcass layer 4, there is provided with an inner liner layer 5. In this case, this inner liner layer 5 is formed from a three layers consisting of an inner layer 5a at the side of the carcass layer 4, an outer layer 5c facing to tire charged air and an intermediate layer 5b to be set up between these layers. And, the outer layer 5a is formed from independent halogenated butyl rubber or its combined natural rubber. And, the intermediate layer 5b is made up of mixing 5W30 wt. of a p-phenylenediamine antioxidant in 100 wt. stock rubber containing less than 20 wt. of natural rubber and/or diene rubber.



LEGAL STATUS

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JAPANESE

[JP,2614441,B]

CLAIMS DETAILED DESCRIPTION TECHNICAL FIELD PRIOR ART EFFECT OF THE INVENTION
DESCRIPTION OF DRAWINGS DRAWINGS

[Translation done.]

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CLAIMS

(57) [Claim(s)]

[Claim 1] the tire which has an inner liner layer inside a carcass layer -- setting -- (1) -- said inner liner layer It consists of three layers with the interlayer stationed between the inner layer by the side of a carcass layer, the outer layer facing tire restoration air, these inner layers, and an outer layer at least. (2) Said outer layer consists of a halogenation isobutylene-isoprene-rubber independent or halogenation isobutylene isoprene rubber, and natural rubber. (3) Said middle class is a pneumatic tire characterized by coming to carry out 5-30 weight section combination of the p-phenylene diamine system antioxidant to the raw material rubber 100 weight section which comes to blend diene system synthetic rubber with a natural rubber independent or natural rubber below 20 weight sections.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[Field of the Invention]

This invention relates to the pneumatic tire which improved endurance sharply, especially the pneumatic tire for heavy loading.

[Description of the Prior Art]

Generally there are two kinds such as the tire with a tube of the type offered with a tube and the tubeless tire of the type offered without a tube of pneumatic tires. Moreover, especially with the tire for heavy loading used for a truck, a bus, etc., since a cap tread is made over repeatedly and long duration use is presented, as compared with the common pneumatic tire, endurance serves as an important military requirement.

In a tire, an inner NAIRA layer is usually arranged inside a carcass layer over the whole inside of a carcass layer. In the case of a tire with a tube, this inner liner layer has the function to protect a carcass layer and a tube, and, in the case of a tubeless tire, it has the function to prevent transparency of the air with which the interior of a tire was filled up. The tire for heavy loading requires especially the function to prevent transparency of moisture in addition to the function of such an inner liner layer, from a demand of high endurance. That is, when a tire is used for a long period of time or it is left by the tire at the time of tire storage, the moisture in air penetrates an inner liner layer. If the metal code is used as a carcass code at this time, an adhesive fall with the powerful fall metallurgy group code and code covering rubber by rust generating of a metal code etc. will take place, and problems, such as cutting of a code, will arise during tire transit. Moreover, even when the organic fiber code of a carcass code is used, wet heat degradation takes place with the transmitted moisture, and it is known better than before that problems, such as cutting of a code, will arise like a metal code. Problems, such as code cutting by this moisture, are problems common to a tire with a tube, and a tubeless tire.

In order to solve these problems, generally the constituent containing non-**** raw material rubber, such as isobutylene isoprene rubber or halogenation isobutylene isoprene rubber, is used for the layer by the side of restoration air from the former among the inner liner layers which consist of two or more layers. However, while a use field expands this kind of tire as compared with the former with an improvement of traffic environments, such as maintenance of a road in recent years, and expansion of a highway, and the demand of the reinforcement of a tire increases further, the demand which improves the endurance of this kind of tire sharply has been increasing. That is, the count of regeneration which comes to be used in the field from a heat-and-high-humidity area to an arctic area, and makes over and uses a cap tread has been increasing. The severe transit conditions in a heat-and-high-humidity area pose a problem especially in the endurance of a tire. In this case, in order to plan the impermeability of restoration air, and the impermeability of moisture, the constituent containing raw material rubber, such as halogenation isobutylene isoprene rubber, is used for the inner liner layer. However, the rubber constituent which used halogenation isobutylene isoprene rubber independently cannot prevent transparency of air and moisture completely, either, and although the approach of thickening thickness of the rubber layer which contains halogenation isobutylene isoprene rubber that transparency of air and moisture should be made as few as possible is also taken, generation of heat of rubber becomes high during transit, and there are problems, such as becoming easy to produce exfoliation by heat deterioration.

[Objects of the Invention]

This invention aims at offering the pneumatic tire equipped with the new inner liner layer which can improve endurance ability sharply in use in a wide range area, holding a function as this kind of a conventional tire.

[Effect of the Invention]

this invention persons found out that the above-mentioned purpose could be attained by limiting the rubber constituent which constitutes at least three layers, and nothing and each class for an inner liner layer, as a result of examining many things.

Therefore, this invention is set into the tire which has an inner liner layer inside a carcass layer. (1) Said inner liner layer consists of three layers with the interlayer stationed between the inner layer and the side of a carcass layer, the outer layer facing tire restoration air, these inner layers, and an outer layer at least. (2) Said outer layer consists of a halogenation isobutylene-isoprene-rubber independent or halogenation isobutylene isoprene rubber, and natural rubber. (3) Said middle class makes a summary the pneumatic tire characterized by coming to carry out 5-30 weight section combination of the p-phenylene diamine system antioxidant to the raw material rubber 100 weight section which comes to blend diene system synthetic rubber with a natural rubber independent or natural rubber below 20 weight sections. Hereafter, the configuration of this invention is explained in detail.

Fig. 1 is the direction half section explanatory view of the meridian of an example of the pneumatic tire of this invention, and Fig. 2 is an expansion explanatory view of the important section.

In Fig. 1, at least one-layer carcass layer 4 is constructed across between the bead wires 3 of a right-and-left pair, and 3, and the belt layer 2 which consists of a steel coat on the carcass layer 4 in a tread 1 is arranged annularly in the tire hoop direction. Inside the carcass layer 4, the whole inside of the carcass layer 4 is covered and the inner liner layer 5 is arranged. 6 is the sidewall section.

The inner liner layer 5 consists of three layers of inner layer 5a by the side of the carcass layer 4, outer layer 5c which faces tire restoration air, and interlayer 5b arranged among these at least, as shown in Fig. 2.

(a) The moisture by which outer layer 5c is contained in restoration air during transparency of air or tire transit, and the moisture in the atmospheric air at the time of tire storage penetrate an inner liner layer. An adhesion fall with the powerful fall metallurgy group code and code covering rubber by rust generating of a metal code when a metal code is used for a carcass code. Since there is similarly a problem of generating, such as a code strengthening fall, on the other hand for wet heat degradation by moisture when the organic fiber code has been arranged in the carcass code, It is required to use the rubber constituent which acts as the subject of the halogenation isobutylene isoprene rubber for the purpose of preventing this, and contains halogenation isobutylene isoprene rubber at least 50% or more among a rubber component (the remainder is natural rubber). Preferably, the rubber constituent which consists of a halogenation isobutylene-isoprene-rubber independent is desirable.

(b) About inner layer 5a and outer layer 5c, although it is two-layer structure in the former, endurance is [in / at this two-layer structure / a heat-and-high-humidity area] dissatisfied by generating of cutting of a carcass code etc. during tire transit. Then, as a result of this invention persons' inquiring wholeheartedly, it became clear that the oxygen in the air which penetrated the inner liner layer was the important cause to rust generating at the time of using a metal code for a carcass layer and the physical-properties fall of carcass code covering rubber. That is, also in generating of rust, only with moisture, a powerful fall could not take place easily and the knowledge that oxidation degradation had broken out by the oxygen in the air which penetrated the physical-properties fall of code covering rubber was acquired. In order to prevent transparency of the oxygen in this air, middle class 5b consists of rubber constituents which carried out 5-30 weight section combination of the p-phenylene diamine system antioxidant to the raw material rubber 100 weight section which comes to blend diene system synthetic rubber with a natural rubber independent or natural rubber below 20 weight sections. If the effectiveness which catches the oxygen in the air which penetrated outer layer 5c when fewer than 5 weight sections is not enough and exceeds 30 weight sections, the effectiveness of preventing transparency of oxygen will cause the physical-properties fall of the tensile strength in early stages of a certain thing etc.

Fig. 3 shows change (O mark in drawing shows.) of the tensile strength of the inner layer rubber when changing the loadings of N-(1, 3-dimethyl-butyl)-N' phenyl-p-phenylene diamine (antioxidant) of middle class rubber among three-layer inner liner layers, and change (- mark in drawing shows.) of the initial tensile strength of middle class rubber. Here, the thickness of a inner layer, an interlayer, and an outer layer is fixed uniformly, and the tensile strength of inner layer rubber is measured by the sample started from the tire after three-week neglect to the 50-degree C thermostat. If the fall of the tensile strength of inner layer rubber is large in under 5 weight sections, and a p-phenylene diamine antioxidant has a problem and exceeds 30 weight sections so that it may become clear from this Fig. 3, the tensile strength in early stages of interlayer rubber falls and is not desirable.

The p-phenylene diamine system antioxidant used in this invention can be chosen from N,N'-diphenyl-p-phenylenediamine, N-phenyl-N'-isopropyl-p-phenylene diamine, N-(1, 3-dimethyl-butyl)-N' phenyl-p-phenylene diamine, N, and N'-screw (1, 3-dimethyl-butyl)-p-phenylene diamine, N, and N'-bis--2-naphthyl-p-phenylene diamine etc. as arbitration. Moreover, in this invention, although optimum dose addition of the compounding agent usually used in the rubber industry other than these compounding agents, for example, a zinc oxide, carbon black, process oil, the vulcanization accelerator, etc. is carried out suitably, it is desirable to choose an accelerator also in it. namely, -- if abundant combination of the antioxidant is carried out -- scorching -- already -- ** -- if a sulfenamide system

vulcanization accelerator is suitable and also blends the loadings in the range of the 0.5 - 1.5 weight section also in the accelerator of thiazoles in order to prevent this since there is fear, scorching is started and the problem of workability can be solved.

The range of the thickness of inner layer 5a 0.5-1.5mm, weight increases by the thickness which there is little effectiveness over stabilization of adhesion with the metal code of carcass code covering rubber, and exceeds 1.5mm in less than 0.5mm, and it is not desirable. Moreover, as for inner layer 5a, it is desirable to use the same constituent as carcass code covering rubber or the rubber constituent near it in order to contact the carcass layer 4 directly. The range of it 0.5-1.0mm, and in order that the thickness of interlayer 5b may blend an antioxidant expensive like the above so much at less than 0.5mm by the thickness which there is little transparency prevention effectiveness of oxygen and exceeds 1.0mm, it is not desirable on economy.

It is good in 1.5-2.5mm, and less than 1.5mm of air and the effectiveness of not penetrating [of moisture] is insufficient, generation of heat becomes high and the thickness of outer layer 5c does not have it on endurance, if 2.5mm is exceeded. [desirable]

Next, an example and the example of a comparison explain this invention in more detail.

An example, example of a comparison With the tire cross-section structure shown in Fig. 1, the radial-ply tire for heavy loading of 1000R20 and the tire size of 11R22.5 was made as an experiment. The carcass layer 4 consists of a steel code, and the code include angle is 90 degrees C of abbreviation to a tire hoop direction. Moreover, the thickness of inner layer 5a, interlayer 5b, and outer layer 5c is 1mm, 1mm, and 2mm in the shoulder region of a tire, respectively.

The rubber constituent of each inner liner layer used for the tire prototype was shown in the 1st table of the following, and the indoor drum endurance life was evaluated in the 2nd table about various kinds of tires using the combination of the rubber constituent shown in the 1st table.

第 1 表

	外層A	内層B	中間層					
			比較例C ₁	実施例C ₂	実施例C ₃	実施例C ₄	比較例C ₅	実施例C ₆
NR	100	100	100	100	100	80	100	100
BR						20		
Br-IIR								
HAF-LS		55	55	55	55	55	55	55
GPF	60							
アロマチックオイル	4	2	2	2	2	2	2	2
ステアリン酸	2	1	1	1	1	1	1	1
亜鉛華	3	8	8	8	8	8	8	8
老化防止剤* ¹	—	2	2	8	20	20	40	20
ナフテン酸コバルト* ²	—	2						2
促進剤DZ* ³	—	0.8	0.8	0.8	0.8	0.8	0.8	0.8
促進剤TT	0.6	—	—					
硫黄* ⁴	1.5	7	7	7	7	7	7	7

注) *¹ N-(1,3ジメチルブチル)-N'フェニル-p-フェニレンジアミン。

*² コバルト元素含有量10%。

*³ N,N'-ジシクロヘキシルベンゾチアジルスルフェンアミド。

*⁴ 油展不溶性イオウ(正味80%重量%)。

第 2 表

	比較例タイヤ1	比較例タイヤ2	実施例タイヤ1	実施例タイヤ2	実施例タイヤ3	実施例タイヤ4
外層	A	A	A	A	A	A
中間層	C ₁	C ₅	C ₂	C ₃	C ₄	C ₆
内層	B	B	B	B	B	B
耐久寿命						
11R22.5 14PR	100	105	120	135	130	140
1000R20 14PR	100	110	130	150	145	155

After leaving an evaluation tire for one month in a 80-degree C thermostat, indoor drum endurance life evaluation measured the transit time until a tire breaks on condition that rate; 50 kg/h internal pressure; 7.25 kgf/cm² and 150% of a load; JIS standard load, and indicated by the characteristic. At this time, all destruction of a tire is destruction of the steel carcass code in the shoulder section.

Each example tire of this invention has the outstanding endurance ability so that clearly from the engine-performance comparison of the 2nd table.

[Effect of the Invention]

As explained above, in the inner liner layer three-tiered structure of this invention, the endurance ability in the wide range operating area from the usual tire use area to a heat-and-high-humidity area can be satisfied. For this reason, especially the tire of this invention is suitable as a pneumatic tire for heavy loading.

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TECHNICAL FIELD

[Field of the Invention]

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PRIOR ART

[Description of the Prior Art]

Generally there are two kinds such as the tire with a tube of the type offered with a tube and the tubeless tire of the type offered without a tube of pneumatic tires. Moreover, especially with the tire for heavy loading used for a truck, a bus, etc., since a cap tread is made over repeatedly and long duration use is presented, as compared with the common pneumatic tire, endurance serves as an important military requirement.

In a tire, an inner NAIRA layer is usually arranged inside a carcass layer over the whole inside of a carcass layer. In the case of a tire with a tube, this inner liner layer has the function to protect a carcass layer and a tube, and, in the case of a tubeless tire, it has the function to prevent transparency of the air with which the interior of a tire was filled up. The tire for heavy loading requires especially the function to prevent transparency of moisture in addition to the function of such an inner liner layer, from a demand of high endurance. That is, when a tire is used for a long period of time or it is left by the tire at the time of tire storage, the moisture in air penetrates an inner liner layer. If the metal code is used as a carcass code at this time, an adhesive fall with the powerful fall metallurgy group code and code covering rubber by rust generating of a metal code etc. will take place, and problems, such as cutting of a code, will arise during tire transit. Moreover, even when the organic fiber code of a carcass code is used, wet heat degradation takes place with the transmitted moisture, and it is known better than before that problems, such as cutting of a code, will arise like a metal code. Problems, such as code cutting by this moisture, are problems common to a tire with a tube, and a tubeless tire.

In order to solve these problems, generally the constituent containing non-**** raw material rubber, such as isobutylene isoprene rubber or halogenation isobutylene isoprene rubber, is used for the layer by the side of restoration air from the former among the inner liner layers which consist of two or more layers. However, while a use field expands this kind of tire as compared with the former with an improvement of traffic environments, such as maintenance of a road in recent years, and expansion of a highway, and the demand of the reinforcement of a tire increases further, the demand which improves the endurance of this kind of tire sharply has been increasing. That is, the count of regeneration which comes to be used in the field from a heat-and-high-humidity area to an arctic area, and makes over and uses a cap tread has been increasing. The severe transit conditions in a heat-and-high-humidity area pose a problem especially in the endurance of a tire. In this case, in order to plan the impermeability of restoration air, and the impermeability of moisture, the constituent containing raw material rubber, such as halogenation isobutylene isoprene rubber, is used for the inner liner layer. However, the rubber constituent which used halogenation isobutylene isoprene rubber independently cannot prevent transparency of air and moisture completely, either, and although the approach of thickening thickness of the rubber layer which contains halogenation isobutylene isoprene rubber that transparency of air and moisture should be made as few as possible is also taken, generation of heat of rubber becomes high during transit, and there are problems, such as becoming easy to produce exfoliation by heat deterioration.

[Objects of the Invention]

This invention aims at offering the pneumatic tire equipped with the new inner liner layer which can improve endurance ability sharply in use in a wide range area, holding a function as this kind of a conventional tire.

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EFFECT OF THE INVENTION

[Effect of the Invention]

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Therefore, this invention is set into the tire which has an inner liner layer inside a carcass layer. (1) Said inner liner layer consists of three layers with the interlayer stationed between the inner layer by the side of a carcass layer, the outer layer facing tire restoration air, these inner layers, and an outer layer at least. (2) Said outer layer consists of a halogenation isobutylene-isoprene-rubber independent or halogenation isobutylene isoprene rubber, and natural rubber. (3) Said middle class makes a summary the pneumatic tire characterized by coming to carry out 5-30 weight section combination of the p-phenylene diamine system antioxidant to the raw material rubber 100 weight section which comes to blend diene system synthetic rubber with a natural rubber independent or natural rubber below 20 weight sections. Hereafter, the configuration of this invention is explained in detail.

Fig. 1 is the direction half section explanatory view of the meridian of an example of the pneumatic tire of this invention, and Fig. 2 is an expansion explanatory view of the important section.

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The inner liner layer 5 consists of three layers of inner layer 5a by the side of the carcass layer 4, outer layer 5c which faces tire restoration air, and interlayer 5b arranged among these at least, as shown in Fig. 2.

(a) The moisture by which outer layer 5c is contained in restoration air during transparency of air or tire transit, and the moisture in the atmospheric air at the time of tire storage penetrate an inner liner layer. An adhesion fall with the powerful fall metallurgy group code and code covering rubber by rust generating of a metal code when a metal code is used for a carcass code, Since there is similarly a problem of generating, such as a code strengthening fall, on the other hand for wet heat degradation by moisture when the organic fiber code has been arranged in the carcass code, It is required to use the rubber constituent which acts as the subject of the halogenation isobutylene isoprene rubber for the purpose of preventing this, and contains halogenation isobutylene isoprene rubber at least 50% or more among a rubber component (the remainder is natural rubber). Preferably, the rubber constituent which consists of a halogenation isobutylene-isoprene-rubber independent is desirable.

(b) About inner layer 5a and outer layer 5c, although it is two-layer structure in the former, endurance is [in / at this two-layer structure / a heat-and-high-humidity area] dissatisfied by generating of cutting of a carcass code etc. during tire transit. Then, as a result of this invention persons' inquiring wholeheartedly, it became clear that the oxygen in the air which penetrated the inner liner layer was the important cause to rust generating at the time of using a metal code for a carcass layer and the physical-properties fall of carcass code covering rubber. That is, also in generating of rust, only with moisture, a powerful fall could not take place easily and the knowledge that oxidation degradation had broken out by the oxygen in the air which penetrated the physical-properties fall of code covering rubber was acquired. In order to prevent transparency of the oxygen in this air, middle class 5b consists of rubber constituents which carried out 5-30 weight section combination of the p-phenylene diamine system antioxidant to the raw material rubber 100 weight section which comes to blend diene system synthetic rubber with a natural rubber independent or natural rubber below 20 weight sections. If the effectiveness which catches the oxygen in the air which penetrated outer layer 5c when fewer than 5 weight sections is not enough and exceeds 30 weight sections, the effectiveness of preventing transparency of oxygen will cause the physical-properties fall of the tensile strength in early stages of a certain thing

etc.

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The range of the thickness of inner layer 5a 0.5-1.5mm, weight increases by the thickness which there is little effectiveness over stabilization of adhesion with the metal code of carcass code covering rubber, and exceeds 1.5mm in less than 0.5mm, and it is not desirable. Moreover, as for inner layer 5a, it is desirable to use the same constituent as carcass code covering rubber or the rubber constituent near it in order to contact the carcass layer 4 directly.

The range of it 0.5-1.0mm, and in order that the thickness of interlayer 5b may blend an antioxidant expensive like the above so much at less than 0.5mm by the thickness which there is little transparency prevention effectiveness of oxygen and exceeds 1.0mm, it is not desirable on economy.

It is good in 1.5-2.5mm, and less than 1.5mm of air and the effectiveness of not penetrating [of moisture] is insufficient, generation of heat becomes high and the thickness of outer layer 5c does not have it on endurance, if 2.5mm is exceeded. [desirable]

Next, an example and the example of a comparison explain this invention in more detail.

An example, example of a comparison With the tire cross-section structure shown in Fig. 1, the radial-ply tire for heavy loading of 1000R20 and the tire size of 11R22.5 was made as an experiment. The carcass layer 4 consists of a steel code, and the code include angle is 90 degrees C of abbreviation to a tire hoop direction. Moreover, the thickness of inner layer 5a, interlayer 5b, and outer layer 5c is 1mm, 1mm, and 2mm in the shoulder region of a tire, respectively.

The rubber constituent of each inner liner layer used for the tire prototype was shown in the 1st table of the following, and the indoor drum endurance life was evaluated in the 2nd table about various kinds of tires using the combination of the rubber constituent shown in the 1st table.

第 1 表

	外層A	内層B	中間層					
			比較例C ₁	実施例C ₂	実施例C ₃	実施例C ₄	比較例C ₅	実施例C ₆
NR	100	100	100	100	100	80	100	100
BR						20		
Br-IIR								
HAF-LS		55	55	55	55	55	55	55
GPF	60							
アロマチックオイル	4	2	2	2	2	2	2	2
ステアリン酸	2	1	1	1	1	1	1	1
亜鉛華	3	8	8	8	8	8	8	8
老化防止剤* ¹	—	2	2	8	20	20	40	20
ナフテン酸コバルト* ²	—	2						2
促進剤DZ* ³	—	0.8	0.8	0.8	0.8	0.8	0.8	0.8
促進剤TT	0.6	—	—					
硫黄* ⁴	1.5	7	7	7	7	7	7	7

- 注) *¹ N-(1,3ジメチルブチル)-N'フェニル-p-フェニレンジアミン。
*² コバルト元素含有量10%。
*³ N,N'-ジシクロヘキシルベンゾチアジルスルフェンアミド。
*⁴ 油展不溶性イオウ(正味80%重量%)。

第 2 表

	比較例タイヤ1	比較例タイヤ2	実施例タイヤ1	実施例タイヤ2	実施例タイヤ3	実施例タイヤ4
外層	A	A	A	A	A	A
中間層	C ₁	C ₅	C ₂	C ₃	C ₄	C ₆
内層	B	B	B	B	B	B
耐久寿命						
11R22.5 14PR	100	105	120	135	130	140
1000R20 14PR	100	110	130	150	145	155

After leaving an evaluation tire for one month in a 80-degree C thermostat, indoor drum endurance life evaluation measured the transit time until a tire breaks on condition that rate;50 kg/h internal pressure;7.25 kgf/cm² and 150% of a load;JIS standard load, and indicated by the characteristic. At this time, all destruction of a tire is destruction of the steel carcass code in the shoulder section.

Each example tire of this invention has the outstanding endurance ability so that clearly from the engine-performance comparison of the 2nd table.

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

Fig. 1 is an explanatory view in which setting the direction half section explanatory view of the meridian of an example of the pneumatic tire of this invention, and Fig. 2 in the expansion explanatory view of the important section, setting Fig. 3 in an inner liner layer, and showing change of the initial tensile strength of an interlayer's antioxidant combination weight section and an interlayer, and the tensile strength of an interlayer's antioxidant combination weight section and a inner layer.

1 [.. A carcass layer, 5 / .. An inner liner layer, 6 / .. The sidewall section, 5a / .. A inner layer, 5b / .. An interlayer, 5c / .. Outer layer.] A tread, 2 .. A belt layer, 3 .. Bead wires, 4

[Translation done.]

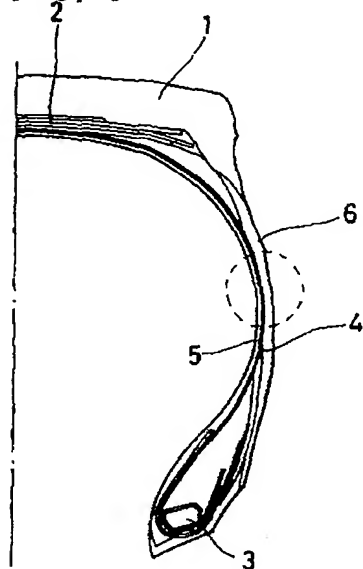
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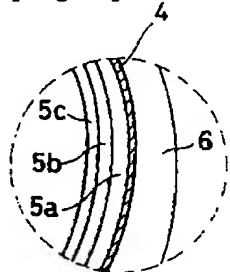
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DRAWINGS

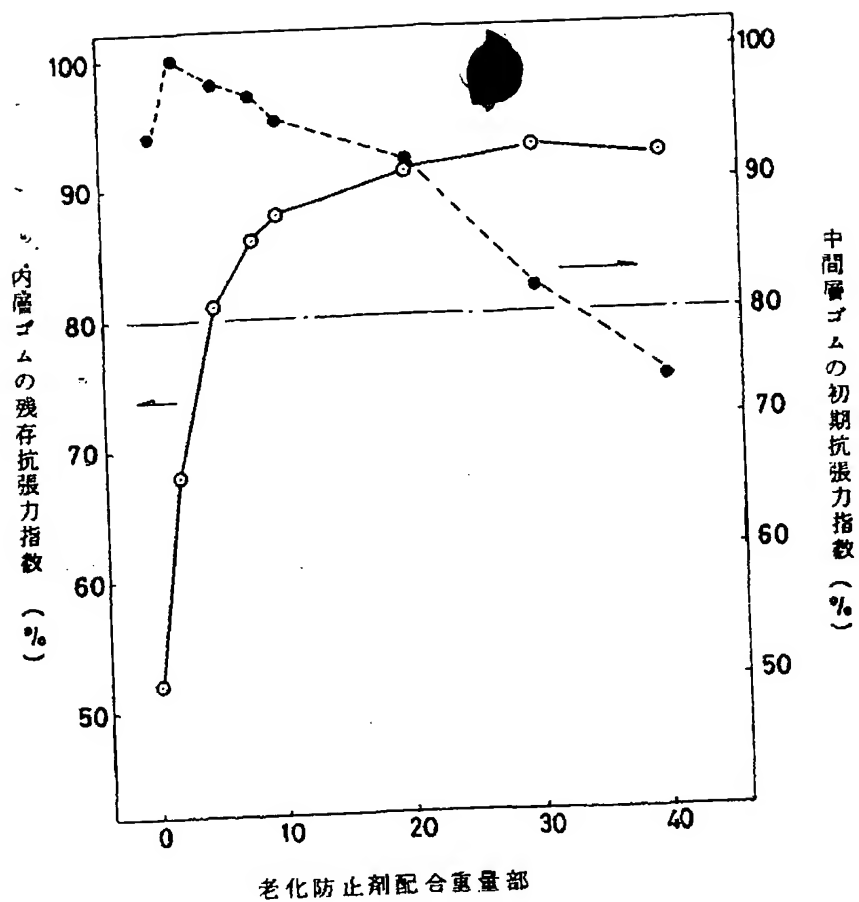
[Fig. 1]



[Fig. 2]



[Fig. 3]



[Translation done.]

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